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CAPTURE-TO-FISSION RATIO MEASUREMENTS OF ^{239}Pu AND ^{241}Pu FROM IRRADIATION OF CAPSULES IN THE EBWR

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by

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December 1971

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ABSTRACT

Planning of an irradiation experiment in the EBWR for the determination of capture-to-fission ratio, α, of plutonium-239 and of plutonium-241 involved hydrodynamic and heat-transfer calculations to ensure that there would be no undue hot spots involved as a result of our experimental arrangement. Our calculations were verified experimentally prior to exposure runs in the reactor.

Mathematical expressions for α^{Pu}, in terms of initial weight measurements and mass-spectrometric and radiochemical data, are given. A discussion of experimental results and of error is also given. Interpolation of our results for bare and cadmium-covered samples to obtain α^{Pu} at thermal energies indicate good agreement with current data at 0.0253 eV.

I. INTRODUCTION

In connection with our study of the buildup of plutonium in boiling-water reactors, the lack of accurate data on α^{49} and α^{41}, especially at epithermal energies, was noted.[1]

In view of our pending Plutonium Recycling Project[2] in the Experimental Boiling Water Reactor (EBWR), it was considered appropriate[3] to carry out some physics experiments by irradiation of capsules of ^{239}Pu and of ^{241}Pu, for the determination of the capture-to-fission ratios, α^{49} and α^{41}, respectively.

It was thought at the time that the integral data obtainable from such irradiation experiments would also serve to check the differential data being obtained.

Our purpose was to indicate the feasibility of carrying out similar research projects in operating reactors by the nuclear industry. Such research could prove to be very beneficial.

Nuclear calculations had indicated that determination of α for capsules set flush on fuel rods would be similar to that of a uniform mixture as in a rod of Pu-U. It was noted from our calculations that determination of α from irradiation of capsules placed in the moderator between fuel rods would be quite different from that of uniform mixtures referred to above. Accordingly, the experimental arrangement shown[3] in Fig. 1 was selected, with a mounting unit as shown in Fig. 2.

A possible objection to our experimental arrangement was that it might cause objectionable hot spots in the fuel elements where the mounting units were set, and that the cadmium covers of our capsules could melt.

Hydrodynamic calculations (see Appendix A) indicated that the mounting assembly could reduce the flow of fluid by as much as 20% in the element surrounding the mounting unit.

To determine the effects of such reductions in fluid flow and of even greater adverse situations, heat-transfer calculations were made (see Appendix B) for fluid-flow reductions as great as 75% and short but thick helium gaps (6-9 mils thick) in a capsule covered with cadmium. It was found that the surface temperature of the fuel rods would not exceed the melting temperature of cadmium (610°C).

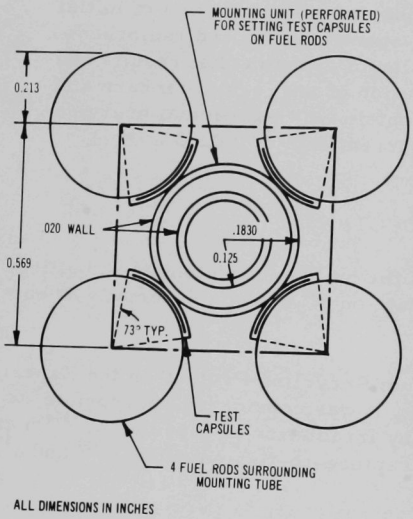

Fig. 1. Experimental Arrangement of Test Capsules Set on Fuel Rods with Aid of Mounting Unit (cross-sectional view shown). ANL Neg. No. 112-6823.

The design of our test capsule (see Fig. 3) was so chosen that in case of any meltdown of cadmium, the plutonium capsule would remain trapped in the specimen holder, as long as the capsule did not fail in any manner, such as by corrosion. The soundness of our design was checked by "autoclave" tests, in which the meltdown temperature of cadmium was exceeded by a factor of 1.5.

Upon completion of the construction and welding of the specimen onto mounting units, the latter were placed in fuel elements Pu-31, Pu-42, E-144, and E-292 of the plutonium and enriched zones (see Fig. 4). There were four mounting units, each carrying four capsules of ^{239}Pu and four of ^{241}Pu, two capsules with cadmium covers and two without.

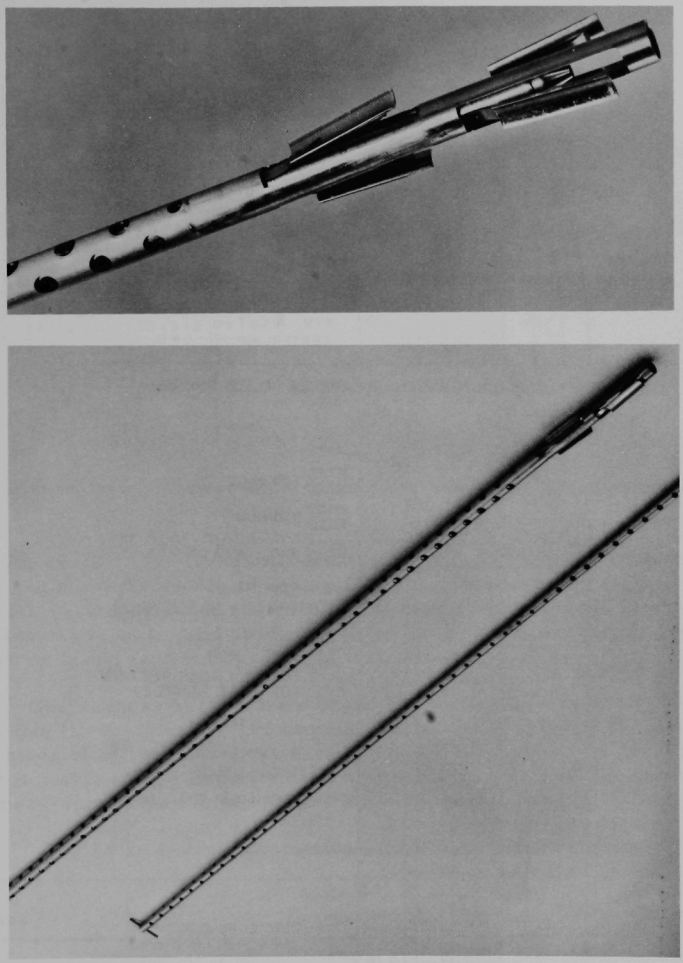

Fig. 2. Mounting-unit Assembly. ANL Neg. Nos. 112-6700 and 112-6701.

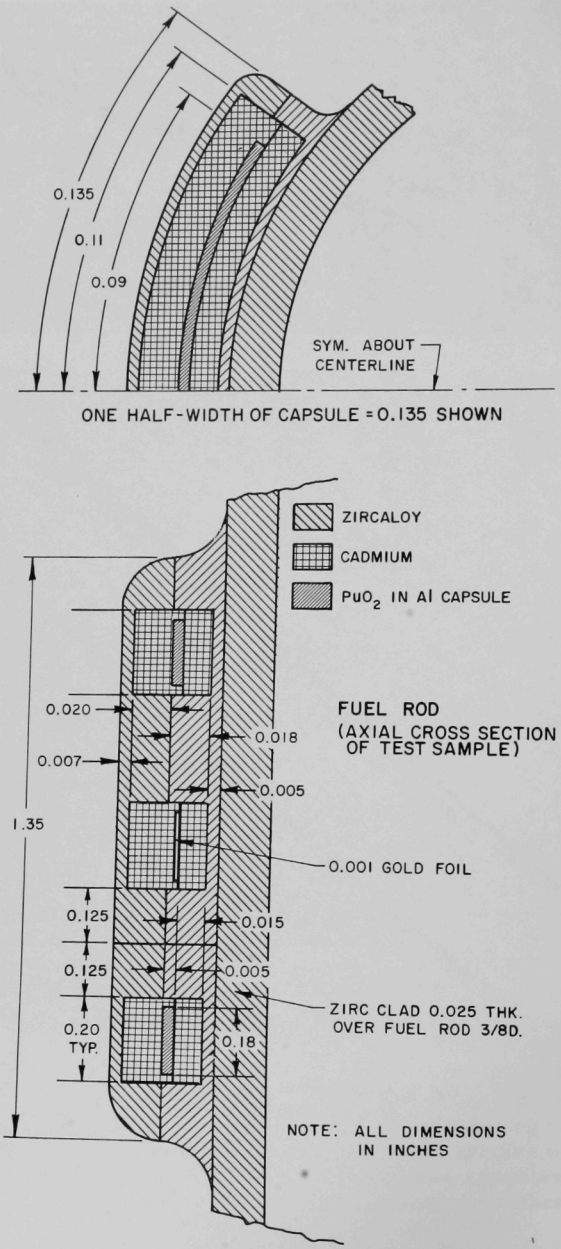

Fig. 3. Cross-sectional View of Test Sample Assembly. ANL Neg. No. 112-6825 Rev. 2.

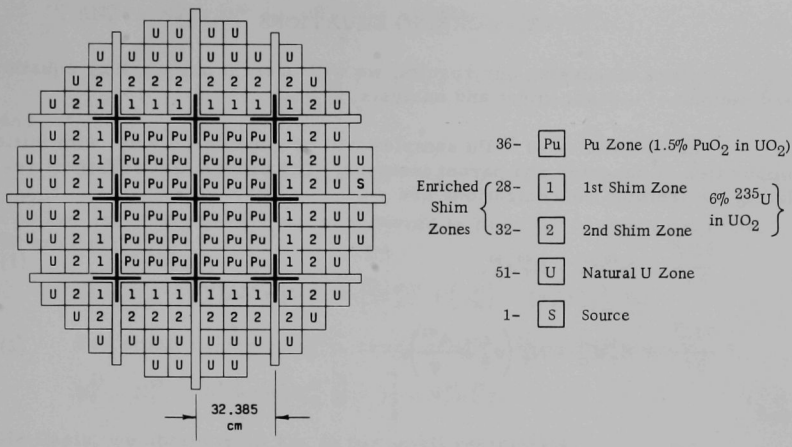

Fig. 4. Schematic Layout of Plutonium Recycle Experiment Core Loading. ANL Neg. No. 112-3630 Rev. 2.

These test specimens were irradiated for 47 days (May 13 to June 29, 1967). After irradiation, the samples were removed from the reactor and allowed to decay to reduce fission-product activity, before the capsules were opened and analyzed. Postirradiation examination showed the capsules to be in good condition. None of the cadmium covers had melted or fused; in fact, they all had their fine edges.

Our samples of ^{239}Pu were of the highest purity obtainable (99.95%; see Table I). Samples of ^{241}Pu were of less purity (see Table I). However, our values of α^{41} are nearly as accurate as for α^{49}, due to the great care taken in making these impurity determinations by radiochemical analysis and mass-spectrometric measurements.

TABLE I. Isotopic Concentration of Unirradiated Plutonium

Mass	Ratio	Mole Percent	Weight Percent	Precision (Sigma)
		^{239}Pu		
239	1.0000000	99.946149	99.945920	0.000331
240	0.0005388	0.053857	0.054082	0.000331
		^{241}Pu		
239	0.0166285	1.514315	1.502247	0.002915
240	0.0680687	6.198822	6.175153	0.006582
241	1.0000000	91.067084	91.097354	0.007018
242	0.0133943	1.219781	1.225249	0.001187

II. WORKING EQUATIONS

Before discussing our results, we will present our working equations and method of measurement and analysis.

The irradiation of ^{241}Pu samples will be considered only, with initial impurities of daughter and parent isotopes. The differential equations relating the isotopic concentrations are

$$\frac{\partial N_\tau^{40}}{\partial \tau} = N_\tau^{49}\sigma_c^{49} - N_\tau^{40}\sigma_a^{40}, \tag{1}$$

$$\frac{\partial N_\tau^{41}}{\partial \tau} = N_\tau^{40}\sigma_c^{40} - N_\tau^{41}\left(\sigma_a^{41} + \frac{\lambda^{41}}{\varphi}\right), \tag{2}$$

and

$$\frac{\partial N_\tau^{42}}{\partial \tau} = N_\tau^{41}\sigma_c^{41} - N_\tau^{42}\sigma_a^{42}, \tag{3}$$

where τ = neutron flux × time (= φt), the subscript τ = 2 refers to concentration of irradiated material at the time of their mass-spectrometric measurements, and λ^{41} = decay constant of ^{241}Pu with T1/2 = 13.2 years. Solutions of Eqs. 1-3 are

$$N_\tau^{40} = N_1^{40}e^{-\sigma_a^{40}\tau} + N_1^{49}\sigma_c^{49}\,\frac{\exp\left(-\sigma_a^{49}\tau\right) - \exp\left(-\sigma_a^{40}\tau\right)}{\sigma_a^{40} - \sigma_a^{49}}, \tag{4}$$

$$N_\tau^{41} = N_1^{41}e^{-\sigma_a^{41'}\tau} + N_1^{40}\sigma_a^{40}\,\frac{\exp\left(-\sigma_a^{40}\tau\right) - \exp\left(-\sigma_a^{41'}\tau\right)}{\sigma_a^{41'} - \sigma_a^{40}}, \tag{5}$$

and

$$N_\tau^{42} = N_1^{42}e^{-\sigma_a^{42}\tau} + N_1^{41}\sigma_c^{41}\,\frac{\exp\left(-\sigma_a^{41}\tau\right) - \exp\left(-\sigma_a^{42}\tau\right)}{\sigma_a^{42} - \sigma_a^{41}}, \tag{6}$$

where the subscript 1 refers to concentration of the unirradiated material at the time of its mass-spectrometric measurements (March 28, 1968), and

$$\sigma_a^{41'} = \sigma_a^{41} + \frac{\lambda^{41}}{\varphi}.$$

Equations 4-6 may be written as

$$\Delta N_\tau^{40} = N_\tau^{40} - N_1^{40} \simeq -N_1^{40}\sigma_a^{40}\tau + N_1^{49}\sigma_c^{49}\tau(1 - \sigma'\tau + \sigma''\tau^2), \quad (7)$$

$$\Delta N_\tau^{41} = N_1^{41} - N_\tau^{41} \simeq N_1^{41}\sigma_a^{41'}\tau - N_1^{40}\sigma_c^{40}\tau(1 - \sigma'\tau + \sigma''\tau^2), \quad (8)$$

and

$$\Delta N_\tau^{42} = N_\tau^{42} - N_1^{42} = N_1^{41}\sigma_c^{41}(1 - \sigma'\tau + \sigma''\tau^2) - N_1^{42}\sigma_a^{42}\tau. \quad (9)$$

For low exposure, the $\sigma'\tau$ and $\sigma''\tau^2$ terms in Eqs. 7-9 are small and may be dropped;

$$\sigma' \equiv \left(\sigma_a^{40} + \sigma_a^{41}\right)/2! \text{ and } \sigma'' \equiv \left[\left(\sigma_a^{40}\right)^2 + \left(\sigma_a^{41}\right)^2 + \left(\sigma_a^{40}\sigma_a^{41}\right)\right]/3!.$$

For small values of $\sigma_a^{41'}\tau$, hence of $\sigma_a^{40}\tau$, we obtain, from Eq. 5,

$$N_2^{41} - N_1^{41} = N_1^{41}\left[\exp\left(-\sigma_a^{41'}\tau\right) - 1\right] + N_1^{40}\sigma_c^{40}\tau. \quad (10)$$

Similarly, we obtain from Eq. 6, for small irradiation,

$$N_2^{42} - N_1^{42} = N_1^{42}\left[\exp\left(-\sigma_a^{42}\tau\right) - 1\right] + N_1^{41}\sigma_c^{41}\tau. \quad (11)$$

Equations 7 and 8 give the loss in ^{241}Pu and gain in ^{242}Pu. We now bring in fission data to obtain α^{41} as follows:

$$\alpha^{41} = \frac{\text{Capture of } ^{241}Pu}{\text{Fission of } ^{241}Pu} = \frac{N_2^{42} - N_1^{42}}{\Delta N_{fiss}^{41}}. \quad (12)$$

We have mass-spectrometric data

$$\left.\begin{aligned} a_1 &= N_1^{42}/N_1^{41} \\ \text{and} \\ a_2 &= N_2^{42}/N_2^{41}. \end{aligned}\right\} \quad (13)$$

Also,

$$N_\tau^{41} \simeq N_1^{41}\exp(-\lambda^{41}\Delta t_1^2) - (1 + \alpha^{41})\Delta N_{fiss}^{41} + N^{40}\sigma_a^{40}\tau, \quad (14)$$

where the factor $e^{-\lambda^{41}\Delta t_1^2}$ allows for decay of ^{241}Pu between the two mass-spectrometric measurements (pre- and postirradiation measurements). The last term of Eq. 11 refers to contribution of ^{240}Pu, which is quite small and therefore will be neglected.

$$\Delta N_{fiss}^{41} = \Delta N_{fiss}^{total} - \Delta N_{fiss}^{49}. \tag{15}$$

Note: Small error will be caused by applying the decay factor to N_1^{41} in Eq. 11, instead of a more accurate value of N^{41}, which allows for fission.

It follows, from Eqs. 9-12, that

$$\alpha^{41} \simeq \frac{a_2[\exp(-\lambda^{41}\Delta t_1^2) - r] - a_1}{(1 + a_2)\, r}, \tag{16}$$

where

$$r = \Delta N_{fiss}^{41}/N_1^{41}. \tag{17}$$

It may similarly be shown for ^{239}Pu, which does not have a decay factor, that

$$\alpha^{49} = \frac{a_2(1 - r) - a_1}{(1 + a_2)\, r},$$

where a_1 and a_2 refer to the preirradiation and postirradiation values of the ratios,

$$a_1 = N_1^{40}/N_1^{49}, \quad a_2 = N_2^{40}/N_2^{49}, \quad \text{and} \quad r = \frac{\Delta N_{fiss}^{49}}{N_1^{49}}. \tag{18}$$

III. METHOD OF MEASUREMENTS

1. Weights of unirradiated test capsules

Activation techniques and initial weighing of total plutonium material yield

$$w_0^{Pu'} = W_0^{Pu} \times a/A$$

where

a = activation of test capsule (α-activation),

A = activation of total mass of plutonium material,

W_0^{Pu} = initial weight of plutonium material,

and

N_1^{49} and N_1^{41} were determined as described in Appendix C.

2. Concentration of isotopes in unirradiated and irradiated capsules

Mass-spectrometric measurements were made as described in Appendix D. For unirradiated samples, see Table I. For irradiated samples, see Tables II-VI. In these tables, the subscript 0 refers to the time of the initial determination of the weight of the samples (February 8, 1967), the subscript 1 refers to the time of the mass-spectrometric measurement of the unirradiated samples (March 28, 1968), the subscript 2 refers to the time of the mass-spectrometric measurement of the irradiated sample, and a_1, a_2, and r are defined by Eq. 18.

3. Purification of capsules of ^{241}Pu for ^{241}Am

This was obtained as indicated in Appendix C.

TABLE II. Nuclear Constants and Experimental Data Used for Determination of α^{41} Bare Samples

Sample from	$w_0^{Pu'}$,[a] μg	N_1^{41}[b] (x 10^{15})	$a_2 = N_2^{42}/N_2^{41}$	ΔN_{fiss}^{total}[c] (x 10^{15})	ΔN_{fiss}^{41}[d] (x 10^{15})	r	Δt_1^2,[e] yr	$e^{-\lambda \Delta t_1^2}$	α^{41}[f]
Pu-31 No. 1	194.04	393.10	0.025615	11.12	10.81	0.02750	0.75	0.9614	0.3727
Pu-31 No. 2	201.85	408.92	0.028036	15.41	15.08	0.03689	0.755	0.9612	0.3298
Pu-42 No. 1	154.72	313.45	0.029638	12.14	11.85	0.0378	1.186	0.9396	0.3426
Pu-42 No. 2	-	-	-	-	-	-	1.186	-	-
		Average =	0.027763		Average =	0.03183		Average =	0.3484
E-144 No. 1	200.51	406.21	0.024120	8.586	8.35	0.02055	1.360	0.9309	0.4067
E-144 No. 2	159.67	323.47	0.025217	8.874	8.63	0.02668	1.360	0.9309	0.3439
E-292 No. 1	217.75	441.14	0.022393	8.252	8.03	0.01822	1.360	0.9309	0.3781
E-292 No. 2	225.05	455.93	0.023557	10.353	10.08	0.02210	1.360	0.9309	0.3543
		Average =	0.023322		Average =	0.02189		Average =	0.3707

[a] $w_0^{Pu'}$ = weight of plutonium (in μg) on February 8, 1967 (not purified).

[b] $N_1^{41} = 2.02 \times 10^{15} \times w_0^{Pu'}$ (see Appendix C).

[c] ΔN_{fiss}^{total} includes fission of ^{239}Pu.

[d] $N_{fiss}^{41} = N^{137}/Y^{137}$ when $Y^{137} = 6.6\%$.

[e] Δt_1^2 = time lapsed between initial mass-spectrometric measurement and final measurement of irradiated capsule.

[f] $\alpha^{41} = \frac{\Delta N^{42}}{N_{fiss}^{41}} = \frac{a_2(e^{-\lambda \Delta t} - r) - a_1}{(1 + a_2) r}$.

TABLE III. Nuclear Constants and Experimental Data[a] Used for Determination of α^{41} Cadmium-covered Samples

Sample from	$W_0^{Pu'}$, µg	N_1^{41} ($\times 10^{15}$)	$a_2 = N_2^{42}/N_2^{41}$	ΔN_{fiss}^{total} ($\times 10^{15}$)	ΔN_{fiss}^{41} ($\times 10^{15}$)[b]	r	Δt_1^2, yr	$e^{-\lambda \Delta t_1^2}$	α^{41}[c]
Pu-31 No. 1	213.07	431.66	0.015893	2.215	2.177	0.005043	0.7473	0.9614	0.3526
Pu-31 No. 2	176.80	358.18	0.016341	2.165	2.133	0.005955	-	0.9614	0.3666
Pu-42 No. 1	199.28	403.72	0.016355	2.045	2.015	0.005066	1.203	0.9396	0.3671
Pu-42 No. 2	174.01	352.5	0.017079	2.803	2.763	0.007840	1.203	0.9396	0.3158
		Average =	0.016517		Average =	0.00597		Average =	0.3505
E-144 No. 1	187.83	380.52	0.016274	1.702	1.67	0.004388	1.3604	0.9311	0.3782
E-144 No. 2	206.53	418.41	0.016606	2.175	2.14[2]	0.005119	1.3604	0.9311	0.3809
E-292 No. 1	161.55	327.28	0.016136	1.517	1.49[2]	0.004558	1.3604	0.9311	0.3360
E-292 No. 2	174.50	353.52	0.016463	1.750	1.72[2]	0.004871	1.3604	0.9311	0.3744
		Average =	0.016120		Average =	0.004734		Average =	0.3674
								Average for all 15 samples =	0.3650

[a] $a_{a1} = N_1^{42}/N_1^{41} = 0.0133943$ for all samples (nonirradiated).

[b] $N_{fiss}^{41} = N^{137}/Y^{137}$ when $Y^{137} = 6.6\%$.

[c] $\alpha^{41} = \frac{\Delta N^{42}}{N_{fiss}^{41}} = \frac{a_2\left(e^{-\lambda \Delta t} - r\right) - a_1}{(1 + a_2) r}$.

TABLE IV. Nuclear Constants and Experimental Data Used for Determination of α^{49} Bare Samples

Sample from	Sample Notation	Plutonium Weight, µg	$N^{49}(0)$ ($\times 10^{15}$)	$N^{40}(0)$[a] ($\times 10^{15}$)	$N_{fiss}^{49}(\tau)$[b] ($\times 10^{15}$)	$a_2 = N_2^{40}/N_2^{49}$	$r = N_{fiss}^{49}(\tau)/N_0^{49}$	α^{49}[c]
Plutonium Zone								
Pu-31 element	EN-12	212.34	534.80	0.2881	15.85	0.01492	0.0297	0.4635
Pu-31 element	EN-11	294.3	741.22	0.3993	28.84	0.01735	0.0389	0.4077
Pu-42 element	EN-14	157.79	397.42	0.2141	-	-	-	-
Pu-42 element	EN-13	212.42	535.01	0.2882	25.3	0.01750	0.0472	0.3356
					Average =	0.01659	Average = 0.0386	Average = 0.4023
Enriched Zone								
EN-144 element	EN-10	297.04	748.12	0.4031	20.33	0.014028	0.0272	0.4637
EN-144 element	EN-09	269.46	678.66	0.3656	18.38	0.012168	0.0272	0.4122
EN-292 element	EN-08	227.44	572.84	0.3070	11.93	0.010440	0.0208	0.4600
EN-292 element	EN-07	146.45	368.85	0.1977	10.20	0.011712	0.0276	0.3880
					Average =	0.01208	Average = 0.0257	Average = 0.4310

[a] $a_{N_{(1)}^{40}} = N_{(0)}^{40} = a_1 N_0^{49}$, where $a_1 = 0.0005338$ for all ^{239}Pu samples determined by mass-spectrometric measurements; $N_2^{40} = a_2 N_2^{49}$, where a_2 was determined by mass-spectrometric measurements.

[b] $N_{fiss}^{49} = N^{137}/Y^{137}$, where $Y^{137} = 6.63\%$ in the fission yield of ^{139}Cs.

[c] $\alpha^{49} = \Delta N^{40}/N_{fiss}^{49} = \frac{a_2(1 - r) - a_1}{(1 + a_2) r}$.

TABLE V. Nuclear Constants and Experimental Data Used for Determination of α^{49} Cadmium-covered Samples

Sample from	Sample Notation	Plutonium Weight, µg	$N^{49}(0)$ ($\times 10^{15}$)	$N^{40}(0)$[a] ($\times 10^{15}$)	$N_{fiss}^{49}(\tau)$[b] ($\times 10^{15}$)	$a_2 = N_2^{40}/N_0^{49}$	$r = N_{fiss}^{49}(\tau)/N_0^{49}$	α_{epi}^{49}[c]
Plutonium Zone								
Pu-31 element	EC-10	243.93	614.36	0.3310	2.551	0.00280	0.00415	0.5400
Pu-31 element	EC-11	196.54	495.3	0.2667	2.351	0.00334	0.00475	0.5850
Pu-42 element	EC-16	243.61	613.58	0.3280	2.503	0.00292	0.00408	0.5800
Pu-42 element	EC-15	233.88	589.07	0.3160	2.8657	0.00326	0.00450	0.5550
					Average =	0.00308	Average = 0.00437	Average = 0.5650
Enriched Zone								
EN-144 element	EC-14	223.57	563.08	0.3034	2.294	0.00281	0.00408	0.5533
EN-144 element	EC-03	212.41	534.97	0.2882	2.068	0.00254	0.00388	0.5155
EN-292 element	EC-07	200.34	504.57	0.2718	1.800	0.00248	0.00357	0.5423
EN-292 element	EC-08	204.51	515.08	0.2775	2.066	0.00285	0.00400	0.5718
					Average =	0.00267	Average = 0.00388	Average = 0.5457

[a] $a_{N_{(1)}^{40}} = N_{(0)}^{40} = a_1 N_0^{49}$, where $a_1 = 0.0005338$ for all ^{239}Pu samples determined by mass-spectrometric measurements; $N_2^{40} = a_2 N_2^{49}$, where a_2 was determined by mass-spectrometric measurements.

[b] $N_{fiss}^{49} = N^{137}/Y^{137}$, where $Y^{137} = 6.63\%$ in the fission yield of ^{139}Cs.

[c] $\alpha^{49} = \Delta N^{40}/N_{fiss}^{49} = \frac{a_2(1 - r) - a_1}{(1 + a_2) r}$.

TABLE VI. Summary of Results for α^{41} and α^{49} and Comparison with Published AERE and IAEA Data

Zone		Bare Sample	Cadmium-covered Sample	Thermal Interpolated Value	AERE Data (2200-m/sec values)		IAEA Data (2200-m/sec values)
$\bar{\alpha}^{41}$	Plutonium	0.3484	0.3505				
$\delta\alpha^{41}_{max}$ [a]	Plutonium	0.0243	0.0360				
a.d.[b]	Plutonium	0.0162	0.0173				
S.D.[c]	Plutonium	0.0220	0.0241				
$\bar{r}^{41}$	Plutonium	0.0340	0.00597	$\bar{\alpha}^{41}_{th}$ = 0.3479	0.390	0.370[d]	0.365
$\bar{\alpha}^{41}$	Enriched	0.3707	0.3674				
$\delta\alpha^{41}_{max}$	Enriched	0.0360	0.0314				
a.d.	Enriched	0.0215	0.0157				
S.D.	Enriched	0.0279	0.0210				
$\bar{r}^{41}$	Enriched	0.0219	0.0473	$\bar{\alpha}^{41}_{th}$ = 0.3716			
$\bar{\alpha}^{41}_{th}$ For all measured samples = 0.3600							
$\bar{\alpha}^{49}$	Plutonium	0.4023	0.5650				
$\delta\alpha^{49}_{max}$	Plutonium	0.0667	0.0250				
a.d.	Plutonium	0.0444	0.0175				
S.D.	Plutonium	0.0642	0.0212				
$\bar{r}^{49}$	Plutonium	0.0386	0.00437	$\bar{\alpha}^{49}_{th}$ = 0.3810			
$\bar{\alpha}^{49}$	Enriched	0.4309	0.5457				0.366
$\delta\alpha^{49}_{max}$	Enriched	0.0430	0.0302				
a.d.	Enriched	0.0309	0.0168				
S.D.	Enriched	0.0370	0.0234				
$\bar{r}^{49}$	Enriched	0.0297	0.0039	$\bar{\alpha}^{49}_{th}$ = 0.4100			

[a] $\delta\alpha_{j_{max}}$ = maximum deviation, from the average, of any sample reading of the group.

[b] a.d. = average deviation.

[c] S.D. = standard deviation.

[d] Refers to data obtained from Chalk River NRX reactor.

IV. DISCUSSION OF RESULTS

It was our intention to determine the capture-to-fission ratios of ^{239}Pu and ^{241}Pu in the neutron spectrum of EBWR for bare and cadmium-covered samples. The accuracies of these values can be estimated from possible errors of measured quantities.

Tables II and III list the nuclear constants and results of mass-spectrometric and radiochemical measurements of irradiated capsules of ^{241}Pu and appropriate decay factors for each sample. These data are given for bare and cadmium-covered samples, irradiated in the plutonium zone and the enriched (6%) shim zone of the EBWR (see Fig. 1). Tables IV and V give similar data for ^{239}Pu.

Table VI summarizes the averages for all test specimens of an isotope as indicated. These are the averages of four capsules of ^{239}Pu and ^{241}Pu irradiated in each zone, except when there was a loss of a capsule, as indicated in Tables II and IV. Table VI also lists thermal values of α obtained from data of bare and cadmium-covered samples, i.e.,

$$\bar{\alpha}_{th} = \frac{\overline{\alpha r}|_{bare} - \overline{\alpha r}|_{Cd\ covd}}{\bar{r}_{bare} - \bar{r}_{Cd\ covd}}.$$

The primary aim of the present work was the determination of α^{49} and α^{41} at integral epicadmium energies in the EBWR. Measurements from bare samples coupled with measurements from cadmium-covered samples would serve, then, to determine α at thermal energies, which could then be compared with presently available data. This method of determining α checked the accuracy of our results.

The last column of Table VI lists the latest IAEA values of α (at 2200-in./sec velocity) and α reactor values given by AERE.

From the data of Tables II-VI, we note the following:

1. The value of α^{49} increases by a factor of about 1.3 to 1.4 with increase in energy from thermal to integrated epithermal.

2. Our values of $\bar{\alpha}^{49}_{th}$ (= 0.381 to 0.410) are for the reactor-hardened spectrum in the EBWR. It is expected that this value should be greater than the α^{49} (2200-m/sec) value because of the hardened spectrum in the EBWR. We note that $\bar{\alpha}^{49}_{epi}$ = 1.3 $\bar{\alpha}^{49}_{th}$ to 1.4 $\bar{\alpha}^{49}_{th}$. The IAEA[6] and the AERE[7] values for ^{239}Pu are at 2200 m/sec (= 0.365).

3. Our values of $\bar{\alpha}^{41}$ show small changes in magnitude over the thermal to integrated epithermal range. Our $\bar{\alpha}^{41}_{th}$ values agree well with the AERE and IAEA values at thermal energies. There are no published integral values of α^{41} at epicadmium energies.

V. CADMIUM RATIO MEASUREMENTS

For spectral information, gold foils with and without cadmium cover were activated during the irradiation of the test samples (see Figs. 1-3).

The ratio of subcadmium to epicadmium absorption was:

In the plutonium zone (1.5% enriched): $\frac{A_{th}}{A_{epi}} = \text{Cd R} - 1 = 0.25.$

In the enriched zone (6% enriched): $\frac{A_{th}}{A_{epi}} = \text{Cd R} - 1 = 0.14.$

Similar data were obtained from radiation measurements of ^{239}Pu and ^{241}Pu (see Table V). From Table V, we have

$$\left.\frac{r_{th}}{r_{epi}} = \frac{(\text{Fiss})_{\text{thermal}}}{(\text{Fiss})_{\text{epith}}}\right|_{\text{Pu zone (1.5\% enr.)}} \qquad \begin{matrix} ^{239}\text{Pu} & ^{241}\text{Pu} \\ = 7.8 & 4.3 \end{matrix}$$

$$\left.\frac{r_{th}}{r_{epi}} = \frac{(\text{Fiss})_{\text{thermal}}}{(\text{Fiss})_{\text{epith}}}\right|_{\text{Enr. zone (6\% enr.)}} \qquad = 5.6 \quad 3.6$$

The above result shows the effect of greater hardness of neutron spectra in the plutonium zone.

VI. ACCURACY OF RESULTS

We may estimate the accuracy of our results from known uncertainties in the measured quantities in our formula for α and from the relative magnitudes of the Standard and Average Deviations of sets of samples.

The quantities involved in the formula for α are (for ^{241}Pu)

$$\alpha^{41} = \frac{a_2(e^{-\lambda\Delta t} - r) - a_1}{(1 + a_2)\, r}. \tag{19}$$

The estimated uncertainties are

$$\frac{\delta a_2}{a_2} = \frac{\delta a_1}{a_1} \simeq 0.005 \quad \text{for mass-spectrometric measurements,}$$

$$\frac{\delta r}{r} \simeq 0.035 \quad \text{allowing for 2\% uncertainties in the value of } Y^{137} \text{ (fission yield of } ^{137}Cs),$$

and

$$\frac{\delta|e^{-\lambda\Delta t}|}{e^{-\lambda\Delta t}} \simeq 0.001 \quad \text{uncertainty in decay of } ^{241}Pu.$$

Our analysis of errors for ^{239}Pu will be the same as that of ^{241}Pu to be given below, except for the omission of the decay-factor term.

We obtain, by differentiation of Eq. 19,

$$\delta\alpha^{41} = \frac{(e^{-\lambda\Delta t} - r)\, a_2 \left|\frac{\delta a_2}{a_2}\right| + a_2 e^{-\lambda\Delta t}\left|\frac{e^{-\lambda\Delta t}}{e^{-\lambda\Delta t}}\right| \pm r\left|\frac{\delta r}{r}\right| \pm \left|\frac{\delta a_1}{a_1}\right| a_1}{(1 + a_2)\, r}$$

$$\pm \frac{[a_2(e^{-\lambda\Delta t} - r) - a_1](1 + a_2)\left|\frac{\delta r}{r}\right| r + r\left|\frac{\delta a_2}{a_2}\right| a_2}{[(1 + a_2)\, r]^2}. \tag{20}$$

To obtain maximum possible errors, + signs are used for

$$\left|\frac{\delta r}{r}\right|, \quad \left|\frac{\delta a_1}{a_1}\right|, \quad \left|\frac{\delta a_2}{a_2}\right|, \quad \text{and} \quad \left|\frac{\delta e^{-\lambda\Delta t}}{e^{-\lambda\Delta t}}\right|.$$

A numerical evaluation of $\delta\alpha^{41}$, Standard Deviation (S.D.), and Average Deviation (a.d.) has been made for each set of data in Table VI,

for samples with and without cadmium covers. Barring cross-contamination of isotopes, our errors are due mainly to uncertainties in the values of a_1, a_2, and r, the major contribution being that due to uncertainties in the value of r, or in estimating Y^{137} and impurities in N_i^{41}. These latter errors are fixed and should occur in the same direction (all high or all low) for all samples, resulting in $\bar{\alpha}_{th}^{41}$ similarly high or low. In view of the good agreement between our interpolated α_{th} and experimental available source data, the above errors are believed greater than actual. We may also point out that if $\delta a/a$ and $\delta r/r$ are of opposite sign, the maximum errors will be reduced in magnitude to only about 2%.

APPENDIX A

Effect of Mounting Unit on Coolant Flow Rate in Fuel Channels

Determination of the effects of the mounting unit with its attached test specimens on the flow rate of the channel coolant was of special interest in the design. Of interest also were the effects of possible gaps in the test elements upon the temperatures of the rods, the zircaloy cladding, and the cadmium covers of the test specimen, since too high temperatures can result in damage to the fuel and/or experiment.

A study was made of the effect of the mounting unit on the rate of fluid flow and steam void concentration. Hydrodynamic calculations of a two-phase flow system have shown that the loss of fluid flow due to the insertion of the mounting unit would be about 20%. The steam-void concentration would correspondingly increase by 20%. (Figure 1 depicts the fuel rod and mounting with test units.) The following is a description of our calculations for EBWR, which is a natural-circulation system.

Bernoulli's (modified form) equation was used in the hydrodynamic calculations:[4]

$$\frac{1}{\rho}\,\Delta P = N\,\frac{v^2}{2g} + (1 - \bar{\alpha})\,L,$$

where

$$N = \frac{fL}{3D}\left[1 + \frac{1 - \chi}{1 - \alpha_e} + \left(\frac{1 - \chi}{1 - \alpha_e}\right)^2\right] + \frac{2\alpha_e}{1 - \alpha_e},$$

$f \equiv$ Moody friction factor,

$D \equiv$ hydraulic diameter $\simeq 4$ x flow area of coolant/wetted perimeter of duct,

$g \equiv$ acceleration due to gravity,

$L \equiv$ channel (boiling zone) length,

$N \equiv$ number of velocity heads undergoing pressure change,

$v \equiv$ inlet coolant velocity,

$\bar{\alpha} \equiv$ mean steam-void fraction,

$\alpha_e \equiv$ exit void fraction,

$\Delta P \equiv$ pressure drop in channel,

$\rho \equiv$ density of coolant,

and

$\chi \equiv$ exit quality of flow (lb of steam/lb of mixture).

The last term, $2\alpha_e/(1 - \alpha_e)$, in the above equation is an approximation of the momentum pressure drop. Since $\chi \ll 1$, this term may be dropped.

Also, a good approximation for this calculation is: $v\alpha$ before insertion of mounting $\simeq v\alpha$ after insertion of mounting.

Values of parameters and constants were as follows:

Before insertion of the mounting unit in an outer element of the plutonium zone,

$$fL/3D = 0.5685,$$

$$v = 3.4 \text{ ft/sec},$$

$$\bar{\alpha} = 0.22,$$

and

D = the hydraulic diameter $= (4A_s/P_w)$,

where A_s = flow area for fuel rod $= (0.569)^2 - (\pi/4)(0.426)^2 = 0.18224$ sq in.; P_w = wetted perimeter $= \pi(0.426) = 1.3383$ in. Hence, $D = 0.5447$ in. $= 0.0454$ ft.

After insertion of the mounting unit, the outer cylinder OD = 0.366 in., ID = 0.326 in.; the inner-cylinder OD = 0.25 in., ID = 0.21 in. Thus,

$$A_s = 0.18224 - \frac{\pi}{4}\left[(0.366)^2 + (0.25^2 - 0.21^2)\right]$$

$$= 0.1500$$

and

$$P_w = \pi[0.426 + (0.346 + 0.23)] = 3.1478.$$

Hence,

$$D = \frac{4 \times 0.1500}{3.1478} = 0.1906 \text{ in.} = 0.01588 \text{ ft}$$

$fL/3D = 0.8485.$

The following were calculated from the above equations and constants:

$v = 2.7$ ft/sec

and

$\bar{\alpha} = 0.265.$

From the above values of v and $\bar{\alpha}$, it was seen that, as stated above, the insertion of the mounting in the channel would cause a reduction of about 20% in flow velocity (and a corresponding increase of 20% in steam void). In EBWR, a natural-circulation system, a reduction in fluid velocity would result in a corresponding local increase in void concentration, retaining a constancy of $v\alpha$; i.e., $v\alpha$ before insertion $\simeq v\alpha$ after insertion.

Comparable effects are expected for the mounting unit inserted in an element at the middle of the enriched shim zone.

APPENDIX B

Effects of Reduced Flow of Fluid and of Gaps in Test Samples

The effect of a possible helium gap 0.00312 in. thick, completely surrounding a long fuel pin, would be to raise the pin's temperature by about 200°F. In our test units with small sample lengths, the temperature rises should, naturally, be much smaller.

To estimate the degree of control required to permit reduction of the gaps in a test specimen and to estimate the combined effect of reduced coolant flow and helium gaps in creating hot spots, three-dimensional THTB[5] calculations were made.

The maximum temperatures at the centerline of the fuel rod, of the zircaloy cladding, and of the cadmium covers were determined under varying conditions of fluid flow and contact resistances (see Table VII).

TABLE VII. Maximum Temperatures of Zircaloy Cladding and Cadmium Covers of Test Unit for Different Flow Rates of Coolant, with and without Gaps in the Test Specimens

T(Centerline of fuel rod) ≈900°F at 40-MW operation
≈1176°F at 70-MW operation

	T, °F			
	40-MW Power Level		70-MW Power Level	
	Cladding	Cadmium	Cladding	Cadmium
Full Flow Rate around Fuel Rods and Test Specimens				
Zero gap between components of test specimen, including cladding	541	510	572	532
0.00312-in. helium gap between outer zircaloy wall and cadmium cover	550	545	614	588
0.00624-in. helium gap between outer zircaloy wall and cadmium cover	-	-	633	609
0.00936-in. helium gap between outer zircaloy wall and cadmium cover	-	-	643	617
75% Flow Rate around Test Specimen, Full Flow over Remaining Area				
Zero gap between components of test specimen, including cladding	542	512	584	534
0.00312-in. helium gap between outer zircaloy wall and cadmium cover	551	547	618	590
0.00624-in. helium gap between outer zircaloy wall and cadmium cover	556	558	636	610
0.00936-in. helium gap between outer zircaloy wall and cadmium cover	557	563	645	619
50% Flow Rate around Half of Rod Covering Test Specimen, Full Flow over Remaining Area				
Zero gap in specimen and rod	545	515	584	540
0.00312-in. helium gap between outer zircaloy wall and cadmium cover	553.5	548.5	693	593
0.00624-in. helium gap between outer zircaloy wall and cadmium cover	-	-	640	614
0.00936-in. helium gap between outer zircaloy wall and cadmium cover	-	-	648	622
25% Flow Rate around Half of Rod Covering Test Specimen, Full Flow over Remaining Area				
Zero gap in specimen and rod	551	522	594	552
0.00312-in. helium gap between outer zircaloy wall and cadmium cover	560	553	628	600
0.00624-in. helium gap between outer zircaloy wall and cadmium cover	-	-	646	620
0.00936-in. helium gap between outer zircaloy wall and cadmium cover	-	-	654	629

Calculations made for 40- and 70-MW power operations indicated no temperature rise sufficient either to melt the cadmium covers or overheat the fuel rods under some adverse conditions of flow rate and/or gap (see Table VII).

.ment, there were 10 test samples each containing cap- and gold, with cadmium covers, and 14 samples without cadmium covers. After the test samples were removed from the reactor, they were opened and examined. In all the test samples, it was ascertained that there had been no fusion of cadmium cups with covers and that the cadmium pieces closely retained their sharp edges. The insertion of the four mounting assemblies in four fuel elements would not change the core pressure in our natural-circulation system.

Summarizing our results, we have found by hydrodynamic calculations that the mounting assembly used in our experiment would reduce the fluid flow by about 20%. Heat-transfer calculations referring to much more adverse conditions, i.e., flow reduction as high as 75% and short gaps as thick as 6-9 mils, indicated that the surface temperature of fuel rods would not exceed the melting temperature of cadmium. Experimental data referring to the actual fuel elements used in the plutonium zone and the enriched zone of EBWR indicated, as expected, that in no case had the cadmium covers placed on or near the surface of the fuel rods reached their melting point of 610°F.

APPENDIX C

Calculations of N_1^{41} in Terms of Weight of ^{241}Pu Which Has Impurities of ^{241}Am in Amount of 7% of ^{241}Pu

Let $W_0^{Pu'}$ = weight of plutonium on February 8, 1967 (not purified)

$$= (W_0^{49} + W_0^{40} + W_0^{42}) + 1.07W_0^{41},$$

where the factor 1.07 allows for impurity of ^{241}Am in the samples. If samples had been chemically cleaned,

$$W_0^{Pu''} = (W_0^{49} + W_0^{40} + W_0^{42}) + W_0^{41}$$

$$= W_0^{Pu} + W_0^{41}.$$

Therefore

$$W_0^{Pu} = W_0^{Pu'} - 1.07W_0^{41}.$$

During the first mass-spectrometric measurements (preirradiated material),

$$W_1^{Pu} = W_0^{Pu} + W_0^{41}e^{-\lambda \Delta t} = W_0^{Pu'} - 0.128W_0^{41},$$

where

$$\Delta t = 3/28/68 - 2/8/67 = 1.1383 \text{ yr}$$

and

$$e^{-\lambda \Delta t} = e^{-0.0596^{\circ}} = 0.9420.$$

Thus,

$$W_1^{Pu} = W_0^{Pu'} - \frac{0.128}{0.942}W_1^{41}.$$

For preirradiated material,

$$W_1^{41} = 0.910973W_1^{Pu},$$

$$W_1^{Pu} + \frac{0.128 \times 0.910973}{0.9420} W_1^{Pu} = W_0^{Pu'},$$

$$W_1^{Pu} = 0.889W_0^{Pu'},$$

$$735 \times 0.889 W_0^{Pu'} = 0.81 W_0^{Pu'},$$

and

$$N_1^{41} = \frac{0.6023}{241} \times W_1^{41} = 2.02 W_0^{Pu'} \times 10^{15},$$

where $W_0^{Pu'}$ is in micrograms. We also have

$$W_0^{Pu} = W_0^{Pu'} - 0.07 W_0^{41} = 0.9398 W_0^{Pu'}.$$

APPENDIX D

Sample Processing: Chemical and Counting Techniques

The radioactivity readings of individual mounting units several days after removal from the reactor ran 10-15 R/hr at 2 in. distance. The center section, which contained the gold flux-monitoring foil, was cut out with a mechanical shear, and the gold removed from the zircaloy cladding for immediate counting of the 2.8-day ^{198}Au activity. The plutonium specimens were put into temporary storage to permit the short-lived fission products to decay out.

1. Gold Flux-monitoring Foils

The bare gold foils were separated readily from the mounting unit and counted without difficulty using a calibrated GeLi gamma-ray detector. The cadmium-wrapped foils, however, had bonded to the cadmium envelope by interfacial alloying. Electron-microprobe studies demonstrated that no significant diffusion into the cadmium foil had occurred, so the cadmium was dissolved away from the gold using 1.8N HNO_3. The recovered gold was somewhat spongy in appearance, but weight measurements indicated that essentially no gold was lost in the cadmium dissolution step, and no ^{115}Cd activity was observed in the counting of the separated gold sponge.

2. Determination of Number of Fissions

The cold-welded aluminum disc containing the ^{239}Pu or ^{241}Pu was mounted on a counting card and analyzed for ^{137}Cs using a 20-cm^3 GeLi detector. Sufficient counts were accumulated for each sample to give standard deviations of 0.5-0.8%, for the 662-keV line of ^{137m}Ba. A set of detector calibration cards was made by preparing similar cards from aliquots of a ^{137}Cs standard solution (Type RS-137 ASTM) containing 1.72×10^{14} atoms/ml determined by mass-spectrometric measurement. The analyzer live time for each count was measured directly from a precision pulser peak in the gamma-ray spectrum, introduced at the detector preamplifier test input. The thermal-neutron fission yields of 6.74 ± 0.14 and 6.60 ± 0.17 for ^{239}Pu and ^{241}Pu, and of 6.58 ± 0.11 for fast fission of ^{239}Pu reported in INC-1277[8] were used for these calculations. A value for the epicadmium fission yield of ^{241}Pu was not available in the literature, but on the basis of discussion with K. F. Flynn (Chemistry Division), the same value of the fission yield was used for the calculation of the epicadmium contribution to ^{241}Pu fission.

3. Analysis for Impurities

A similar, but unirradiated, ^{241}Pu specimen was dissolved and analyzed by emission spectroscopy to confirm that no significant ($<<1\%$ weight) impurities were present, except for the ^{241}Am daughter. The weight

:ach specimen was determined by GeLi counting of the omparison to a ^{241}Am standard calibrated by the International Atomic Energy Agency. The number of ^{241}Pu atoms present in each sample at the time of irradiation was calculated from the corrected sample weight and the measured isotopic composition.

4. Preparation of Samples for Mass-spectrometric Measurements

The PuO_2 sample was dissolved, and an aliquot containing approximately 10 μg of plutonium was purified by ion-exchange chromatography using a 6 x 60-mm column of Dowex-1 (100-200 mesh) resin in the NO_3 form. The plutonium was absorbed from 8N HNO_3 feed solution. The measured decontamination factor for ^{241}Am in this procedure is >100. The purified plutonium was eluted with 0.1N HNO_3 for analysis on the mass spectrometer.

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